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Diffusion of Inert and Hydrogen-Bonding Solutes in Aliphatic Alcohols

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The diffusion of various inert and of hydrogen-bonding solutes into aliphatic alcohols was considered. The diffusivities of benzene, *p*-dichlorobenzene, biphenyl, and triphenyl methane in methanol, ethanol, *n*-propanol, *n*-butanol, *n*-pentanol, and *n*-heptanol were measured at 25°C. for low solute concentrations. An equation was developed for the limiting values of the binary mutual diffusion coefficient in complex-forming mixtures. This equation, used in conjunction with the Wilke-Chang correlation, gave a considerable improvement over existing predictive methods for the diffusivity of inert solutes, mono- and polyhydroxy alcohols, and water in aliphatic alcohols.

This work is concerned with diffusion of inert solutes (that is, solutes incapable of forming hydrogen bonds, such as carbon tetrachloride) and hydrogen-bonding solutes into aliphatic monohydroxy alcohol solvents. It is part of a wider study of diffusion in dilute complex-forming binary liquid mixtures. The study was undertaken because solute-solvent complexes are formed by hydrogen bonding in many organic solutions, and complex formation often leads to large errors in the diffusivity predicted by the literature correlations (*1* to *8*). Alcohol solvents are of special interest, because they have a high hydrogen-bonding capacity.

Consider, for example, Figure 1, where we have compared experimentally observed diffusivities in methanol and ethanol with values predicted by the Wilke-Chang equation (*1*) for inert solutes, for mono- and polyhydroxy alcohols, and for water. The experimental data were taken from Tables 1, 2, and 3. The association parameters for methanol and ethanol were taken as 1.9 and 1.5, respectively (*1*). From Figure 1 it can be seen that the predicted diffusivities for inert solutes are consistently low, often by more than 20%. On the other hand, the predicted diffusivities for mono- and polyhydroxy alcohols and water are always higher than the observed values. This is so because hydrogen bonding between solute and solvent hydroxyl groups tends to retard the diffusional motion of these latter types of solutes—a factor which is not taken into account in the correlation.

Thus the Wilke-Chang equation, which was developed using data for all types of solutes (*1*), performs best for mixtures where moderate solute-solvent interactions are present. However, where interactions are either fairly weak or very strong, this equation can give large errors in the

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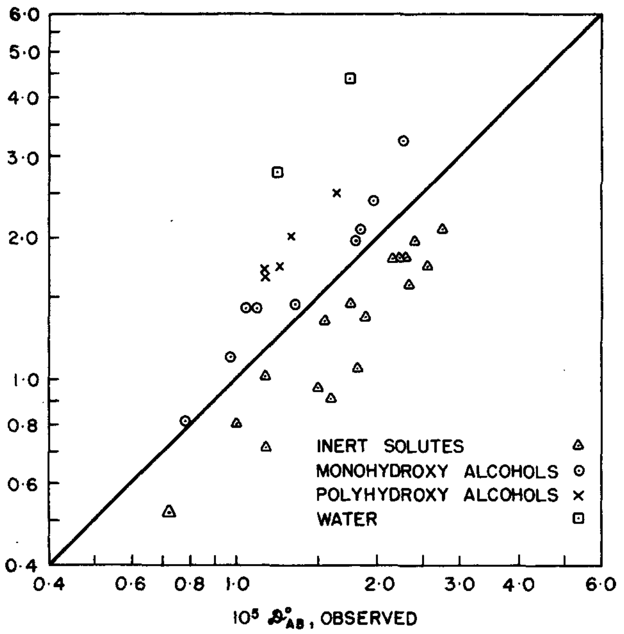

Fig. 1. Comparison of observed diffusivities in methanol and ethanol with values predicted by the Wilke-Chang equation.

predicted diffusivity. Similar trends are observed with other literature correlations (*1* to *8*).

For solvents which can form strong hydrogen bonds, one way of improving earlier predictive methods for the diffusivity might be to take more careful account of the difference between the activation energies for mutual diffusion and viscous flow (*28, 29*). In this work we examine another approach: we consider the nature of the molecular complexes formed, and their diffusional behavior (references *30* and *31*). We shall confine attention to the modifications that such considerations produce in the Wilke-Chang correlation, because it is one of the most frequently used, and has already been examined in past work on the diffusion of water in alcohols (*9, 10*).

THEORY

Consider diffusion in an associated solvent B, such as an aliphatic alcohol, which in the pure state consists of monomers B_1 and complexes $B_2, \ldots B_g \ldots$ (g indicates the number of monomer units in the complex B_g). If the solute A has a tendency to form complexes with the solvent, then at infinite dilution the species present in the binary mixture will be solute and solvent monomers (A_1 and B_1) and various complexes (designated A_1B_g and B_g). Thus the associated binary mixture of A and B can also be viewed as a multicomponent mixture consisting of monomers and complexes (*11*).

Now the molar flux of the solute is related to the experimentally measured diffusion coefficient $\mathcal{D}_{AB}$ by the equation

$$J_A = -\,\mathcal{D}_{AB}\,\nabla C_A \tag{1}$$

When regarded as a multicomponent mixture of monomers and complexes, the multicomponent diffusion equations for species containing solute molecules reduce, at infinite dilution, to

$$J_{A_1} = -\,D_{A_1}\,\nabla C_{A_1} \tag{2}$$

and

$$J_{A_1B_g} = -\,D_{A_1B_g}\,\nabla C_{A_1B_g} \qquad g = 1, 2, \ldots \tag{3}$$

But a mass balance requires that

$$J_A = J_{A_1} + \sum_g J_{A_1B_g} \tag{4}$$

and therefore Equations (1) to (4) give the following expression for the mutual diffusion coefficient, at infinite dilution of the solute:

$$\mathcal{D}^0{}_{AB} = D_{A_1}\frac{dC_{A_1}}{dC_A} + \sum_g D_{A_1B_g}\frac{dC_{A_1B_g}}{dC_A} \tag{5}$$

The derivatives in Equation (5) can be evaluated if we adopt the simple continuous association model for the associated mixture (*11*). This model assumes that all degrees of association are possible for the alcohol solvent ($g = 1, 2, \ldots \infty$), and that the association constants for each of the following two reactions are the same for all values of g:

$$A_1 + B_g \overset{K_{AB}}{\rightleftarrows} A_1B_g \qquad g = 1, 2, \ldots$$

$$B_1 + B_g \overset{K_B}{\rightleftarrows} B_{g+1} \qquad g = 1, 2, \ldots$$

TABLE 1. DIFFUSION OF INERT SOLUTES IN ALCOHOLS

			$10^5\,\mathcal{D}^0{}_{AB}$, sq. cm./sec.	
Solute	Solvent	t, °C.	Observed	Equation (14) (with the present association parameters)
Benzene	Methanol	27	2.76 (*15*)	2.74
Toluene		25	2.56 (*16*)	2.28
Biphenyl		25	1.89	1.78
Triphenyl methane		25	1.16	1.33
Carbon tetrachloride		25	2.25 (*17*)	2.40
Carbon tetrachloride		25	2.30 (*18*)	2.40
Ethyl bromide		15	2.40 (*19*)	2.57
Ethyl iodide		15	2.16 (*19*)	2.38
Bromobenzene		15	1.75 (*19*)	1.91
p-Dibromobenzene		15	1.55 (*19*)	1.75
p-Dichlorobenzene		25	2.35	2.10
Benzene	Ethanol	25	1.81 (*20*)	1.56
Toluene		15	1.60 (*21*)	1.35
Biphenyl		25	1.16	1.05
Triphenyl methane		25	0.718	0.776
Carbon tetrachloride		25	1.50 (*18*)	1.42
Iodobenzene		20	1.01 (*19*)	1.18
Benzene	*n*-Propanol	25	1.28	1.25
Toluene		25	1.35 (*22*)	1.08
Biphenyl		25	0.727	0.844
Triphenyl methane		25	0.394	0.640
p-Dichlorobenzene		25	1.05	0.991
Benzene	*n*-Butanol	25	0.988	1.07
Biphenyl		25	0.627	0.719
p-Dichlorobenzene		25	0.817	0.845
Benzene	*n*-Pentanol	25	0.985	0.911
Biphenyl		25	0.571	0.615
p-Dichlorobenzene		25	0.733	0.724
Methyl cyclohexane	*n*-Heptanol	25	0.470 (*23*)	0.470
Benzene		25	0.64	0.59
p-Dichlorobenzene		25	0.475	0.469

TABLE 2. DIFFUSION OF MONOHYDROXY ALCOHOLS IN ALCOHOLS

Solute	Solvent	t, °C.	$10^5\, \mathcal{D}^0{}_{AB}$, sq. cm./sec.	
			Observed	Equations (16) and (17)
Methanol	Methanol	25	2.27 (*24*)	2.11
Allyl alcohol		15	1.80 (*19*)	1.55
i-Propanol		30	1.97 (*25*)	1.95
i-Butanol		30	1.84 (*25*)	1.74
i-Amyl alcohol		15	1.34 (*19*)	1.28
Ethanol	Ethanol	25	1.01 (*24*)	1.05
Ethanol		25	1.05 (*26*)	1.05
Allyl alcohol		20	0.96 (*19*)	0.89
i-Amyl alcohol		20	0.78 (*19*)	0.74
i-Propanol	*i*-Propanol	25	0.649 (*24*)	0.675
Methanol	*n*-Propanol	30	0.80 (*25*)	0.91
n-Propanol		25	0.646 (*24*)	0.685
i-Butanol		30	0.58 (*25*)	0.70
Methanol	*i*-Butanol	30	0.59 (*25*)	0.58
n-Propanol		30	0.40 (*25*)	0.51
n-Butanol	*n*-Butanol	25	0.504 (*24*)	0.505

TABLE 3. DIFFUSION OF WATER AND POLYHYDROXY ALCOHOLS IN ALCOHOLS

Solute	Solvent	t, °C.	$10^5\, \mathcal{D}^0{}_{AB}$, sq. cm./sec.	
			Observed	Equations (16) and (17)
Water	Methanol	15	1.75 (*10*)	1.75
Water	Ethanol	25	1.22 (*10*)	1.03
Water	*n*-Propanol	15	0.61 (*10*)	0.51
Water	*i*-Propanol	15	0.38 (*10*)	0.52
Water	*n*-Butanol	25	0.56 (*10*)	0.54
Water	*i*-Butanol	15	0.30 (*10*)	0.30
Ethylene glycol	Methanol	25	1.64 (27)	1.64
Glycerol	Methanol	15	1.15 (*19*)	1.22
Glycerol	Methanol	25	1.31 (27)	1.48
Propylene glycol	Methanol	15	1.24 (*19*)	1.28
Erythritol	Methanol	25	1.15 (27)	1.40

These assumptions give the following distributions of complex sizes [the relationships below are valid for infinite dilution of the solute, and may be readily derived from the equations given in Chapter 26 of reference (*11*)]:

$$C_{A_1B_g} = K_{AB}\, C_{A_1}\, C_{B_g} \tag{6}$$

and

$$C_{B_g} = K_B{}^{g-1}\, C^g{}_{B_1} \tag{7}$$

A mass balance on the solute requires that

$$C_A = C_{A_1} + \sum_{g=1}^{\infty} C_{A_1B_g} \tag{8}$$

After substituting Equations (6) and (7) into Equation (8), and closing the resulting infinite geometrical progression, we obtain the following relationship between C_A and C_{A_1}:

$$C_A = \left[1 + \frac{K_{AB}}{K_B} \left(\frac{K_B C_{B_1}}{1 - K_B C_{B_1}} \right) \right] C_{A_1} \tag{9}$$

where $K_B C_{B_1}$, the product of the solvent association constant and monomer concentration, is a characteristic quantity for each alcohol solvent. Substituting Equations (6) and (7) into Equation (9), one also obtains

$$C_A = \frac{K_B}{K_{AB}} \frac{1}{(K_B C_{B_1})^g} \left[1 + \frac{K_{AB}}{K_B} \left(\frac{K_B C_{B_1}}{1 - K_B C_{B_1}} \right) \right] C_{A_1B_g} \tag{10}$$

Equations (9) and (10) may be used to evaluate the derivatives in Equation (5). The final result is

$$\mathcal{D}^0{}_{AB} = \frac{D_{A_1}}{\left[1 + \frac{K_{AB}}{K_B} \left(\frac{K_B C_{B_1}}{1 - K_B C_{B_1}} \right) \right]} \left\{ 1 + \frac{K_{AB}}{K_B} \sum_{g=1}^{\infty} \frac{D_{A_1B_g}}{D_{A_1}} (K_B C_{B_1})^g \right\} \tag{11}$$

Equation (11) indicates that if there is no association between the solute and solvent ($K_{AB} = 0$), the limiting value of the mutual diffusion coefficient is, as one would expect, simply equal to the diffusivity of the solute monomer in the solvent. This is the case for inert solutes. If, however, a solute-solvent complex is formed, then the monomer diffusivity is no longer equal to the mutual diffusion coefficient.

For the latter case, we shall assume that D_{A_1} and $D_{A_1B_g}$ can be estimated (using the correct molar volume in each case) from the Wilke-Chang equation, as modified for the diffusion of inert solutes in alcohols (see below). This is permissible because the associated mixture is here viewed as a multicomponent solution of monomers and complexes. From this viewpoint only normal intermolecular forces exist between the species in the mixture (*11*), which is also the case for inert solutes. Hence solute monomers, and complexes containing solute molecules, diffuse through the solvent in the same way as inert solute molecules.

Using the Wilke-Chang equation to estimate D_{A_1} and $D_{A_1B_g}$, and noting that the LeBas molar volume of species A_1B_g is

$$V_{A_1B_g} = V_A + gV_B$$

one may write Equation (11) in the form

$$\mathcal{D}^0{}_{AB} = \frac{D_{A_1}}{\left[1 + \frac{K_{AB}}{K_B} \left(\frac{K_B C_{B_1}}{1 - K_B C_{B_1}} \right) \right]} \left[1 + \frac{K_{AB}}{K_B} f \left(\frac{V_B}{V_A}, K_B C_{B_1} \right) \right] \tag{12}$$

where

$$f \left(\frac{V_B}{V_A}, K_B C_{B_1} \right) = \sum_{g=1}^{\infty} \frac{(K_B C_{B_1})^g}{\left[1 + g \left(\frac{V_B}{V_A} \right) \right]^{0.6}} \tag{13}$$

and D_{A_1} is the diffusivity of solute monomers, as predicted by the Wilke-Chang equation for inert solutes.

EXPERIMENT

As noted, the original Wilke-Chang correlation (*1*) does not predict well the diffusivities of inert solutes in methanol and ethanol. Therefore we decided to find better values of the association parameters in this correlation, using data already published along with experimental results of our own. As a guide in choosing solutes, we used the classification of liquids proposed by Ewell et al. (*12*). Thus all solutes belonging to their class V are incapable of forming hydrogen bonds with the solvent, and are by our definition inert.

We used a diaphragm cell technique described elsewhere (*13*) to obtain diffusion coefficients, at 25°C., for benzene, *p*-dichlorobenzene, biphenyl, and triphenyl methane in methanol, ethanol, *n*-propanol, *n*-butanol, *n*-pentanol, and *n*-heptanol. Experiments for each system were repeated two or three times, and the standard deviation of our results was usually less than 3% of the reported diffusivity. The measured diffusivities were essentially limiting values, because the solute concentration was always very small (less than 0.5 mole %).

Our results for the diffusion of inert solutes, along with those of other workers, are shown in Table 1. To check the accuracy of our experimental method, we examined the diffusion of carbon tetrachloride in methanol and ethanol, obtaining diffusivity values of 2.28×10^{-5} and 1.52×10^{-5} sq.cm./sec., respectively. These agree well with the published values (Table 1).

Table 2 shows the diffusivities of monohydroxy alcohols in alcohol solvents, and Table 3 the values for water and polyhydroxy alcohol solutes. The diffusivities of water shown in Table 3 are those Lees and Sarram (*10*) consider to be the most reliable.

DISCUSSION OF RESULTS

The value of the Wilke-Chang association parameter χ_B, defined by the equation

$$\frac{\mathfrak{D}^0_{AB}\,\mu_B}{T} = 7.4 \times 10^{-8}\,\frac{\sqrt{\chi_B M_B}}{V_A^{0.6}} \qquad (14)$$

was previously found to be 1.9 for methanol and 1.5 for ethanol (*1*). For the higher alcohols some authors have used a value of 1.5 (*14*), while others have assumed smaller values of χ_B (*10*).

It was found that the data shown in Table 1 for the diffusion of inert solutes in methanol and ethanol can be represented, with an average error of 10%, by Equation (14) with χ_B equal to 3.3. The data for propanol, butanol, pentanol, and heptanol can be correlated, with an average error of 7% (not including the system triphenyl methane-propanol), with χ_B equal to 5.1.

The values of the association parameter found here for methanol and ethanol are different from those reported by Wilke and Chang (*1*), because we have considered the diffusivities of inert solutes only, whereas a large proportion of the data correlated by Wilke and Chang came from systems where solute-solvent complexes are formed. Also, the fact that χ_B is larger for the higher alcohols than for methanol and ethanol suggests that, even for the diffusion of inert solutes, the association parameter is not related to the degree of association of the solvent only (*1*). If this were the case, then one would have expected methanol and ethanol, which have a greater proportion of hydroxyl groups (and hence probably a higher degree of association), to have the larger value of χ_B.

Consider now the diffusion of solutes which form complexes with alcohol solvents. The simplest case is that of self-diffusion, where the solute is simply an isotope of the solvent. Here

$$\frac{V_B}{V_A} = \frac{K_{AB}}{K_B} = 1 \qquad (15)$$

and thus it is easy to estimate, for each solvent, the characteristic value $K_B C_{B_1}$ in Equation (12). The self-diffusion coefficients for methanol through butanol were used for this purpose. It was found that $K_B C_{B_1}$ has a value of 0.83 ± 0.03 for all the alcohols. Using this average value of $K_B C_{B_1}$, one reduces Equation (12) to

$$\mathfrak{D}^0_{AB} = \frac{D_{A_1}}{\left[1 + 4.9\,\frac{K_{AB}}{K_B}\right]}\left[1 + \frac{K_{AB}}{K_B}\,f\left(\frac{V_B}{V_A}, 0.83\right)\right] \qquad (16)$$

where D_{A_1} is given by Equation (14) with χ_B equal to 3.3 for methanol and ethanol and 5.1 for higher alcohols. The function $f\left(\frac{V_B}{V_A}, 0.83\right)$, obtained from a computer evaluation of Equation (13), is shown (for all the cases of interest here) in Figure 2. Although the constants in Equation (16) were evaluated from data at 25°C. only, the equation was found to be useful for other temperatures as well.

For cases other than self-diffusion, the following assumption was made about the ratio K_{AB}/K_B:

$$\frac{K_{AB}}{K_B} = (n_{OH})^{5.2} \qquad (17)$$

where n_{OH} is the number of hydroxyl groups in the solute. For water n_{OH} was assumed to be equal to 1.5. The exponent on n_{OH} was obtained by a back-calculation, using Equation (16) and the single experimental diffusivity for the water-methanol system shown in Table 3.

According to Equation (17), K_{AB} and K_B are equal if both solute and solvent are monohydroxy alcohols. This is a reasonable first approximation. If the solute is a polyhydroxy alcohol, then one would expect the larger number of hydroxyl groups on the solute molecule to increase the hydrogen-bonding tendency between solute and solvent. Equation (17) indicates this, although other simple assumptions (for example, that the ratio of association constants is proportional to the number of solute hydroxyl groups) gave results almost as good.

Diffusivities predicted by Equations (16) and (17) for monohydroxy alcohol solutes gave an average error of 8% when compared with the experimental values shown in Table 2. For the diffusion of water into alcohols, Equations (16) and (17) gave results comparable to Olander's water-as-tetramer model (*9*, *10*), the average error being 12%. Good results were also found for the diffusion of polyhydroxy alcohol solutes. Diffusivities for the systems shown in Table 3 were predicted with an average error of 9%.

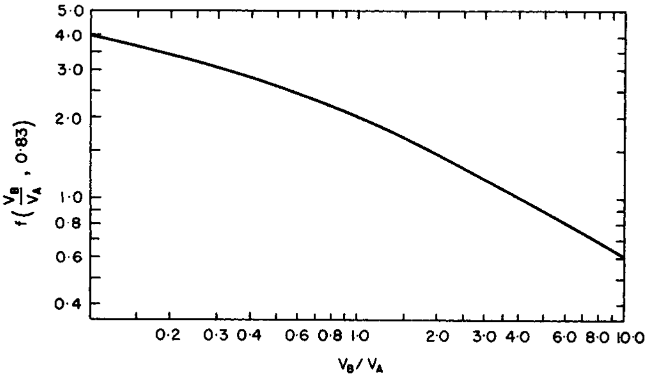

Fig. 2. Value of the function $f(\frac{V_B}{V_A}, 0.83)$ in Equation (16).

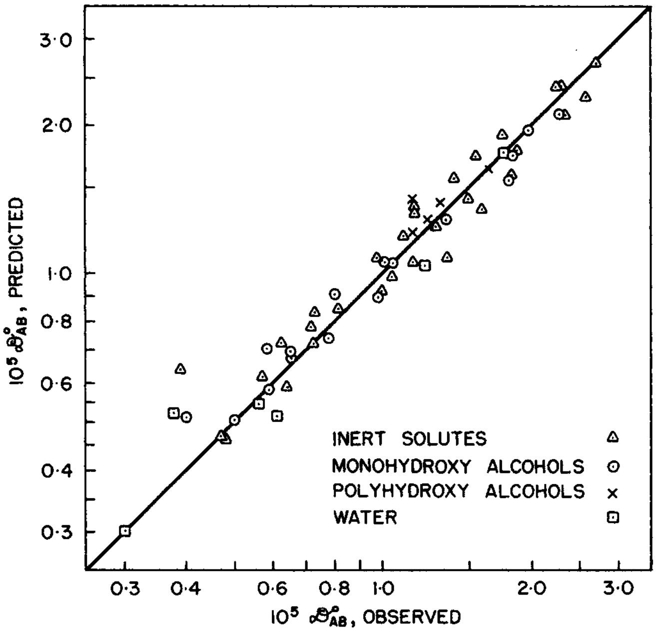

Fig. 3. Comparison of observed diffusivities in *n*-monohydroxy aliphatic alcohol solvents with values predicted by Equation (16).

The diffusivities calculated using Equations (14) to (17) are plotted against the observed values, for the systems shown in Tables 1 to 3, in Figure 3. This figure may be compared with Figure 1. It can be seen that a considerable improvement has been effected by taking special account of solute-solvent complex formation. The majority of the predicted diffusivities are now within 15% of the observed values. By means of Equation (16), the results for the diffusion of inert solutes, mono- and polyhydroxy alcohols, and water have been brought into a unified scheme.

RECOMMENDATIONS

The following recommendations can be made for predicting diffusion coefficients in monohydroxy alcohol solvents:

1. If the solute is inert, K_{AB} in Equation (16) is equal to zero. To predict the diffusivity, use Equation (14) with χ_B equal to 3.3 for methanol and ethanol and 5.1 for the higher alcohols.
2. For monohydroxy alcohol solutes, use Equation (16), with K_{AB} equal to K_B.
3. For water and polyhydroxy alcohol solutes, use Equations (16) and (17).

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NOTATION

C_A = stoichiometric concentration of the solute, g.-moles/cc.
C_{A_1}, $C_{A_1B_g}$ = concentration of solute monomers, solute-solvent complexes, g.-moles/cc.
C_{B_1}, C_{B_g} = concentration of solvent monomers, solvent complexes, g.-moles/cc.
$\mathcal{D}^0_{AB}$ = limiting value of the mutual diffusion coefficient, at infinite dilution of the solute, sq.cm./sec.
D_{A_1}, $D_{A_1B_g}$ = diffusivity of solute monomers, solute-solvent complexes in the solvent, sq.cm./sec.
J = molar flux, g.-moles/(sq.cm.) (sec.)
K_{AB}, K_B = association constant for solute-solvent, solvent-solvent complex formation, cc./g.-mole
M_B = molecular weight of the solvent
n_{OH} = number of hydroxyl groups in the solute
T = absolute temperature, °K.
V_A, V_B = LeBas molar volume (*14*) of solute, solvent cc./g.-mole
χ_B = solvent association parameter in Equation (14)
μ_B = solvent viscosity, centipoises

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